

# Configuration interaction method for out-of-equilibrium correlated many-body systems – admixing nonequilibrium quasiparticle excitations to density matrix

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## Abstract.

We develop configuration interaction method to treat far from equilibrium quantum many-body systems. The concept of embedding (buffer zones between the reservoirs and the correlated quantum system) is used to derive an exact master equation for the reduced density matrix. To enable the use of field theoretic methods we employ the superoperator techniques in Liouville-Fock space. Within the suggested method the exact steady-state density matrix is represented as a linear combination all possible nonequilibrium quasiparticle excitations built on the appropriate reference state. As an example we consider the electron transport through the system with electron-phonon interaction. Using approximate (truncated) expansion of the density matrix we obtain the linear system of equations for two-quasiparticle amplitudes. Then we compute the current and compare the result with other approaches. The current conserving property of the method is proved.

## 1. Introduction

The description of correlated quantum many-body systems far from equilibrium remains one of the challenging problems in modern statistical mechanics, quantum field theory, nuclear, atomic, molecular and condensed matter physics.[1, 2, 3] Most of theoretical approaches are based on Keldysh nonequilibrium Green's functions (NEGF) which enables perturbative treatment of nonequilibrium many-body systems.[4, 3] NEGF also allows systematic summation of specific classes of nonequilibrium diagrams, for example, random phase approximation[5] or GW theory.[6, 7] Nonperturbative methods, such as, numerical renormalization group theory, are also available but often restricted to the oversimplified model systems.[8] In this paper we propose a new theoretical method – nonequilibrium configuration interaction (NECI), which provides a nonperturbative treatment and, in principle, is able to achieve the exact solution of the nonequilibrium problem by representing the density matrix as a linear combination of nonequilibrium quasiparticle excitations above the vacuum state.

Let us briefly explain the main ideas of our approach. We start from the superoperator representation of the master equation for the density matrix. [9, 10, 11, 12]. A summary of the superoperator formalism relevant to this work is provided in Appendix A, while a detailed discussion can be found elsewhere [9, 13]. In this representation nonequilibrium dynamics of many-body system is given by the Schrödinger-like equation

$$i \frac{\partial}{\partial t} |\rho(t)\rangle = L |\rho(t)\rangle \quad (1)$$

where  $|\rho(t)\rangle$  is the vector in Liouville-Fock space, which corresponds to the density matrix, and  $L$  is the Liouville superoperator (Liouvillian). In this paper, we focus on the nonequilibrium steady-state density matrix, which is a solution of the equation

$$L|\rho\rangle = 0. \quad (2)$$

Once the equation is solved, the average of any physical operator  $A$  can be computed according to

$$\langle A \rangle = \text{Tr}[A\rho] = \langle I | \hat{A} | \rho \rangle. \quad (3)$$

Here,  $\langle I |$  is the bra-vector in the Liouville-Fock space which corresponds to the unit operator  $I$ , and the superoperator  $\hat{A}$  is related to an operator  $A$  according to definition (A.4). Thus, our goal is to find the Liouville-Fock space vector which satisfies condition (2).

By the application of Wick theorem in Liouville-Fock space[14] the Liouvillian can be brought to the normal ordered form. The diagonalization of the quadratic part in terms of nonequilibrium quasiparticles, i.e. in terms of normal modes of the uncorrelated part of the Liouvillian, yields:[14, 15, 13]

$$\begin{aligned} L &= \sum_n (\Omega_n \hat{c}_n^\dagger \hat{c}_n - \Omega_n^* \tilde{c}_n^\dagger \tilde{c}_n) + L'(\hat{c}^\dagger, \hat{c}, \tilde{c}^\dagger, \tilde{c}) \\ &= L^{(0)} + L', \end{aligned} \quad (4)$$

where  $\hat{c}^\dagger$ ,  $\tilde{c}^\dagger$  and  $\hat{c}$ ,  $\tilde{c}$  are creation and annihilation superoperators for nonequilibrium quasiparticles. The Liouvillian (4) describes a system of nonequilibrium quasiparticles with complex spectrum  $\Omega_n$ ,  $-\Omega_n^*$  and the quasiparticle-quasiparticle interaction  $L'$ . [15, 13]

Following the ideas of equilibrium configuration interaction method, [16] in order to describe many-body correlations we need to specify a reference state with respect to which the correlations are defined. We choose the reference state as a vacuum for nonequilibrium quasiparticles, i.e.,  $\hat{c}_n |\rho^{(0)}\rangle = \tilde{c}_n |\rho^{(0)}\rangle = 0$ . Then the exact nonequilibrium density matrix can be represented a linear combination of the vacuum, two nonequilibrium quasiparticle excitations, four nonequilibrium quasiparticle excitations and so on. Therefore, NECI expansion for the correlated exact density matrix has the following form

$$|\rho\rangle = (1 + S) |\rho^{(0)}\rangle, \quad (5)$$

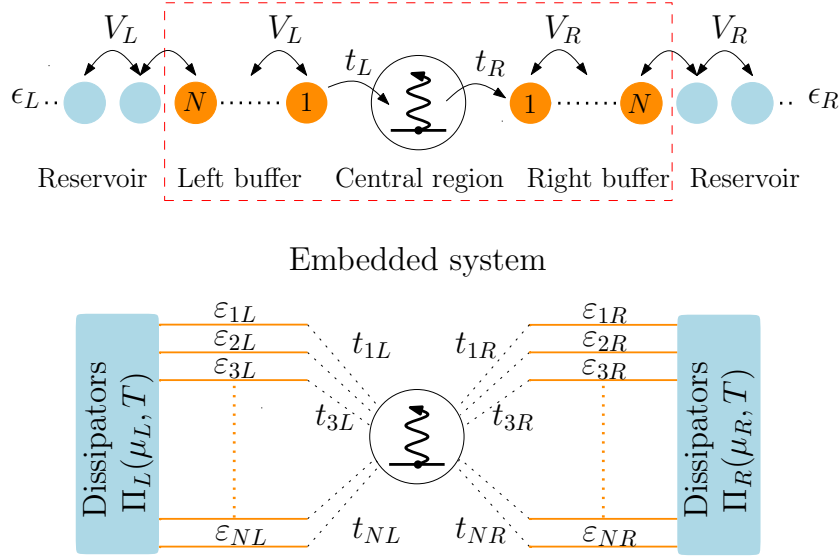
where  $S = \sum_k S_k$  contains a sum of the multi-quasiparticle creation superoperators which generate  $k$ -quasiparticle excitations above the reference state  $|\rho^{(0)}\rangle$ , i.e.,  $S_2 \sim \hat{c}_m^\dagger \tilde{c}_n^\dagger$ ,  $S_4 \sim \hat{c}_m^\dagger \hat{c}_n^\dagger \tilde{c}_p^\dagger \tilde{c}_q^\dagger$ , etc. The relative weights (amplitudes) of multi-quasiparticle excitations can be found by demanding that the density matrix (5) satisfies the steady-state condition (2), i.e.

$$LS |\rho^{(0)}\rangle = -L' |\rho^{(0)}\rangle \quad (6)$$

This equation is equivalent to the system of inhomogeneous linear equations for the amplitudes of the density matrix expansion. To compute the average of a single-particle operator (current, population, etc.) we need to know two-quasiparticle amplitudes. However, some truncation are to be made in the density matrix expansion for practical purposes. This results in an approximate solution of equation (6).

In this paper, we develop this approach on the specific example of electron transport through a quantum system (central region) with electron-phonon interaction. But the method can be easily extended to an arbitrary correlated central region.

The remainder of the paper is organized as follows. In Sec. 2, we introduce the Lindblad master equation for an embedded system and its superoperator representation. Section 3 presents the nonequilibrium configuration interaction method. We discuss two different truncated expansions for the density matrix and apply the theory to calculate the steady-state current. In this section, we also proof that NECI method is current conserving. The numerical NECI calculations of the steady-state current and comparison with other approaches are presented in Sec. 4. Conclusions are given in Sec. 5. Appendix A contains a summary of the superoperator formalism. Finally, in Appendix B we provide details about NEGF calculations within the first Born approximation and the second-order perturbation theory. We use natural units throughout the paper:  $\hbar = k_B = |e| = 1$ , where  $-|e|$  is the electron charge.



**Figure 1.** Schematic illustration of embedding of an open, interacting quantum system. Upper part: Each electrode is divided into a macroscopic reservoir and a finite buffer zone. The region enclosed by the red dashed line is the embedded system. Lower part: The projection of the reservoirs results into the Lindblad master equation for the reduced density matrix of the embedded quantum system. Each buffer zone energy level  $\varepsilon_{k\alpha}$  is connected to the dissipator and the central region.

## 2. Lindblad master equation in Liouville-Fock space

As shown in Figure 1, we consider a paradigmatic model of nonequilibrium many-body theory: the interacting quantum system (central region) connected to two noninteracting electrodes, left ( $L$ ) and right ( $R$ ), maintained with different chemical potentials,  $\mu_L$  and  $\mu_R$ , and the same temperature  $T$ . To be specific, we focus on the electron transport problem through one electronic single-particle level with energy  $\varepsilon_0$  coupled linearly to a vibrational mode (phonon) of frequency  $\omega_0$  (the so called local Holstein model). Thus, the central region Hamiltonian is

$$H_S = \varepsilon_0 a^\dagger a + \omega_0 d^\dagger d + \kappa a^\dagger a (d^\dagger + d), \quad (7)$$

where  $a^\dagger$  ( $a$ ) and  $d^\dagger$  ( $d$ ) are electron and phonon creation (annihilation) operators, respectively. The left and right electrodes are modeled as semi-infinite tight-binding chains of atoms characterized by the hopping matrix element  $V_\alpha$  and the on-site energy  $\varepsilon_\alpha$  ( $\alpha = L, R$ ). The coupling between the central region and the end site of  $\alpha$  electrode is given by the matrix element  $t_\alpha$ .

We replace the infinite system by a finite one using open boundary conditions. To this end each electrode is partitioned into two parts: the macroscopically large reservoir and the finite buffer zone between the central region and the reservoir.<sup>‡</sup> In doing so,

<sup>‡</sup> The idea of the buffer zone was first proposed by us[14, 15, 13] and then also employed by Ajisaka et al.[17]

the Hamiltonian of the whole system takes the form

$$\mathcal{H} = H_S + H_{SB} + H_B + H_{BR} + H_R. \quad (8)$$

In the energy representation the reservoir and the buffer zone Hamiltonians are diagonal

$$H_R = \sum_{l\alpha} \varepsilon_{l\alpha} a_{l\alpha}^\dagger a_{l\alpha}, \quad H_B = \sum_{k\alpha} \varepsilon_{k\alpha} a_{k\alpha}^\dagger a_{k\alpha}, \quad (9)$$

where  $\varepsilon_{l\alpha}$  denote the continuum single-particle spectra of the left ( $\alpha = L$ ) and right ( $\alpha = R$ ) reservoir states, while the buffer zones have discrete energy spectrum  $\varepsilon_{k\alpha}$ . The buffer-reservoir and central region-buffer couplings have the standard tunneling form:

$$H_{BR} = \sum_{lk\alpha} (v_{lk\alpha} a_{l\alpha}^\dagger a_{k\alpha} + \text{h.c.}), \quad (10)$$

$$H_{SB} = \sum_{k\alpha} (t_{k\alpha} a_{k\alpha}^\dagger a + \text{h.c.}). \quad (11)$$

Projecting out the reservoir degrees of freedom from the Liouville-von Neumann equation for the total density matrix, we obtain Lindblad master equation for the reduced density matrix of the embedded system (central region + finite buffer zones) [18, 13]

$$i \frac{\partial \rho(t)}{\partial t} = [H, \rho(t)] + i \Pi \rho(t). \quad (12)$$

Here,  $H$  is the Hamiltonian of the embedded system which includes the Lamb shift of the buffer zone single-particle levels

$$H = H_S + H_{SB} + H_B + \sum_{k\alpha} \Delta_{k\alpha} a_{k\alpha}^\dagger a_{k\alpha}, \quad (13)$$

and  $\Pi \rho(t)$  is the non-Hermitian dissipator given by the standard Lindblad form

$$\Pi \rho(t) = \sum_{k\alpha} \sum_{\mu=1,2} (2L_{k\alpha\mu} \rho(t) L_{k\alpha\mu}^\dagger - \{L_{k\alpha\mu}^\dagger L_{k\alpha\mu}, \rho(t)\}). \quad (14)$$

The operators  $L_{k\alpha 1}$  and  $L_{k\alpha 2}$  are referred to as the Lindblad operators, which represent the buffer-reservoir interaction. They have the following form:

$$L_{k\alpha 1} = \sqrt{\Gamma_{k\alpha 1}} a_{k\alpha}, \quad L_{k\alpha 2} = \sqrt{\Gamma_{k\alpha 2}} a_{k\alpha}^\dagger. \quad (15)$$

with  $\Gamma_{k\alpha 1} = \gamma_{k\alpha}(1 - f_{k\alpha})$ ,  $\Gamma_{k\alpha 2} = \gamma_{k\alpha}f_{k\alpha}$ . Here  $f_{k\alpha} = [1 + e^{(\varepsilon_{k\alpha} - \mu_\alpha)/T}]^{-1}$  and  $\gamma_{k\alpha}$  ( $\Delta_{k\alpha}$ ) is the imaginary (real) part of the self-energy arising from the buffer-reservoir interaction  $\sum_l |v_{lk\alpha}|^2 / (\varepsilon_{k\alpha} - \varepsilon_{l\alpha} + i0^+)$ .

The Lindblad master equation (12) describes the time evolution of an open embedded quantum system preserving Hermiticity, normalization, and positivity of the nonequilibrium density matrix. Open boundary conditions are taken into account by the

non-Hermitian dissipative part,  $\Pi\rho(t)$ , which represents the influence of the reservoir on the buffer zone. The applied bias potential enters into the master equation via fermionic occupation numbers  $f_{k\alpha}$  which depend on the temperature and the chemical potential in the left and right electrodes. We have recently demonstrated that this embedding procedure makes the master equation (12) exact in the steady-state regime, if the buffer zones are large enough to cure the deficiencies of Born-Markov approximation for treating the buffer-reservoir interface.[14, 15, 13]

We use a superoperator formalism and convert the Lindblad master equation (12) to the non-Hermitian Schrödinger-like equation for the nonequilibrium density matrix.[15, 13] Within the superoperator formalism the density matrix is considered as a vector in the Liouville-Fock space and all other operators as superoperators. In Appendix A we present a brief summary of the formalism and demonstrate how to convert the Lindblad master equation into a superoperator form. The resulting equation is

$$i\frac{\partial}{\partial t}|\rho(t)\rangle = (\hat{H} - \tilde{H} - i\sum_{k\alpha}\Pi_{k\alpha})|\rho(t)\rangle = L|\rho(t)\rangle. \quad (16)$$

Here, the superoperators  $\hat{H} = \hat{H}_S + \hat{H}_{SB} + \hat{H}_B$  and  $\tilde{H} = \tilde{H}_S + \tilde{H}_{SB} + \tilde{H}_B$  are obtained from the Hamiltonian (13) by replacing ordinary creation and annihilation operators  $a^\dagger$ ,  $a$  with non-tilde,  $\hat{a}^\dagger$ ,  $\hat{a}$ , and tilde,  $\tilde{a}^\dagger$ ,  $\tilde{a}$ , superoperators, respectively (note that we include the Lamb shift into  $H_B$ ). The dissipators are given by

$$\begin{aligned} \Pi_{k\alpha} = & (\Gamma_{k\alpha 1} - \Gamma_{k\alpha 2})(\hat{a}_{k\alpha}^\dagger \hat{a}_{k\alpha} + \tilde{a}_{k\alpha}^\dagger \tilde{a}_{k\alpha}) \\ & - 2i(\Gamma_{k\alpha 1}\tilde{a}_{k\alpha}\hat{a}_{k\alpha} + \Gamma_{k\alpha 2}\tilde{a}_{k\alpha}^\dagger \hat{a}_{k\alpha}^\dagger) + 2\Gamma_{k\alpha 2}. \end{aligned} \quad (17)$$

The Liouville superoperator  $L$  is non-Hermitian because of  $\Pi_{k\alpha}$ . Some other important properties of  $L$  are discussed in Appendix A. In particular, the relation  $\langle I|L = 0$  holds, where the Liouville-Fock space bra-vector  $\langle I|$  corresponds to the unit operator  $I$ .

We focus our attention on asymptotic ( $t \rightarrow \infty$ ) nonequilibrium steady-state situation, where the density matrix  $|\rho(t)\rangle$  does not depend on time. Therefore the problem is reduced to the problem of finding the right zero-eigenvalue eigenvector of the non-Hermitian finite-dimensional Liouville superoperator

$$L|\rho\rangle = 0. \quad (18)$$

Knowing the solution of (18), we can compute the steady-state current into the central region from  $\alpha$  buffer zone as

$$J_\alpha = \langle I|\hat{J}_\alpha|\rho\rangle, \quad (19)$$

where

$$\hat{J}_\alpha = -i\sum_k t_{k\alpha}(\hat{a}_{k\alpha}^\dagger \hat{a} - \hat{a}^\dagger \hat{a}_{k\alpha}) \quad (20)$$

is the current superoperator.

In the next section we discuss the development of NECI method to solve equation (18). We also present the application of NECI to compute the steady-state current through the central region.

### 3. Configuration interaction method for nonequilibrium density matrix

In the beginning of the section, we make the important remark on the notation use in the rest of the paper: only creation/annihilation superoperators written with letters  $a$ ,  $d$  (such as for example  $\hat{a}_{b\alpha}$  and  $\hat{a}_{b\alpha}^\dagger$ ) are related to each other by the Hermitian conjugation; all other creation  $\hat{c}^\dagger$ ,  $\hat{b}^\dagger$ ,  $\hat{\gamma}^\dagger$  and annihilation  $\hat{c}$ ,  $\hat{b}$ ,  $\hat{\gamma}$  superoperators (as well as their tilde-conjugated partners) are "canonically conjugated" to each other, i.e., for example,  $\hat{c}^\dagger$  does not mean  $(\hat{c})^\dagger$  although  $\hat{c}\hat{c}^\dagger \pm \hat{c}^\dagger\hat{c} = 1$  ( $\pm$  - bosons/fermions).

#### 3.1. Nonequilibrium quasiparticles

Let us introduce nonequilibrium quasiparticle creation and annihilation superoperators, which are the basic building blocks for the NECI method. We start by decomposing the Liouvillian as

$$L = L^{(0)} + L', \quad (21)$$

where  $L^{(0)} = L_{\text{el}}^{(0)} + L_{\text{ph}}^{(0)}$  is the Liouvillian for noninteracting electrons and phonons,

$$\begin{aligned} L_{\text{el}}^{(0)} &= \varepsilon_0(\hat{a}^\dagger\hat{a} - \widetilde{a}^\dagger\widetilde{a}) + (\hat{H}_{SB} + \hat{H}_B - \text{t.c.}) - i \sum_{k\alpha} \Pi_{k\alpha}, \\ L_{\text{ph}}^{(0)} &= \omega_0(\hat{d}^\dagger\hat{d} - \widetilde{d}^\dagger\widetilde{d}), \end{aligned} \quad (22)$$

while  $L'$  represents the interaction between them

$$L' = \kappa\{\hat{a}^\dagger\hat{a}(\hat{d}^\dagger + \hat{d}) - \text{t.c.}\} \quad (23)$$

Hereinafter, the notation 't.c.' stands for tilde conjugated superoperators (see the tilde-conjugation rules in Appendix A).

Nonequilibrium quasiparticles are defined by diagonalizing the electronic part of  $L^{(0)}$ :

$$L_{\text{el}}^{(0)} = \sum_n (\Omega_n \hat{c}_n^\dagger \hat{c}_n - \Omega_n^* \widetilde{c}_n^\dagger \widetilde{c}_n), \quad (24)$$

where  $1 \leq n \leq 2N + 1$ . Using the equation of motion method we find non-unitary (but canonical) Bogoliubov transformation which diagonalizes  $L_{\text{el}}^{(0)}$ :

$$\begin{aligned} \hat{c}_n &= \psi_n \hat{b} + i\varphi_n \widetilde{b}^\dagger + \sum_{k\alpha} (\psi_{n,k\alpha} \hat{b}_{k\alpha}^\dagger + i\varphi_{n,k\alpha} \widetilde{b}_{k\alpha}^\dagger), \\ \hat{c}_n^\dagger &= \psi_n \hat{b}^\dagger + \sum_{k\alpha} \psi_{n,k\alpha} \hat{b}_{k\alpha}^\dagger, \quad \widetilde{c}_n = (\hat{c}_n)^\sim, \quad \widetilde{c}_n^\dagger = (\hat{c}_n^\dagger)^\sim, \end{aligned} \quad (25)$$

where

$$\begin{aligned} \hat{b}^\dagger &= \hat{a}^\dagger - i\widetilde{a}, \quad \hat{b} = \hat{a}, \quad \widetilde{b}^\dagger = (\hat{b}^\dagger)^\sim, \quad \widetilde{b} = (\hat{b})^\sim, \\ \hat{b}_{k\alpha}^\dagger &= \hat{a}_{k\alpha}^\dagger - i\widetilde{a}_{k\alpha}, \quad \hat{b}_{k\alpha} = (1 - f_{k\alpha})\widetilde{a}_{k\alpha} - if_{k\alpha}\widetilde{a}_{k\alpha}^\dagger, \\ \widetilde{b}_{k\alpha}^\dagger &= (\hat{b}_{k\alpha}^\dagger)^\sim, \quad \widetilde{b}_{k\alpha} = (\hat{b}_{k\alpha})^\sim, \end{aligned} \quad (26)$$

Amplitudes  $\psi$ ,  $\varphi$  are the solution of the following systems of equations

$$\begin{aligned}\varepsilon_0 \psi_n - \sum_{k\alpha} t_{k\alpha} \psi_{n,b\alpha} &= \Omega_n \psi_n, \\ E_{k\alpha} \psi_{n,k\alpha} - t_{k\alpha} \psi_n &= \Omega_n \psi_{n,k\alpha},\end{aligned}\tag{27}$$

and

$$\begin{aligned}(\varepsilon_0 - \Omega_n) \varphi_n - \sum_{k\alpha} t_{k\alpha} \varphi_{n,k\alpha} &= \sum_{k\alpha} t_{k\alpha} f_{k\alpha} \psi_{n,k\alpha}, \\ (E_{k\alpha}^* - \Omega_n) \varphi_{n,k\alpha} - t_{k\alpha} \varphi_n &= -t_{k\alpha} f_{k\alpha} \psi_n.\end{aligned}\tag{28}$$

with  $E_{k\alpha} = \varepsilon_{k\alpha} + \Delta_{k\alpha} - i\gamma_{k\alpha}$ . By solving the eigenvalue problem (27) we also obtain the quasiparticle spectrum  $\Omega_n$ ,  $-\Omega_n^*$ .

Taking into account property (A.7), we see that  $\langle I|$  is a vacuum state for  $\hat{c}_n^\dagger$  and  $\tilde{c}_n^\dagger$  superoperators,

$$\langle I| \hat{c}_n^\dagger = \langle I| \tilde{c}_n^\dagger = 0.\tag{29}$$

Although superoperators  $\hat{c}_n^\dagger$  and  $\hat{c}_n$  ( $\tilde{c}_n^\dagger$  and  $\tilde{c}_n$ ) are not Hermitian conjugate to each other, they obey the fermionic anticommutation relations. Particularly, from  $\{\hat{c}_n^\dagger, \hat{c}_n\} = 1$  it follows that  $\psi$  amplitudes are normalized according to

$$(\psi_n)^2 + \sum_{k\alpha} (\psi_{k\alpha})^2 = 1.\tag{30}$$

Other useful relations between amplitudes  $\psi$ ,  $\varphi$  can be derived by making use of the transformation inverse to (25) (see relations (A4) in our previous paper [14]) and computing the anticommutators. For example

$$\begin{aligned}\{\hat{b}, \tilde{b}_{k\alpha}^\dagger\} = 0, &\Rightarrow \sum_n (\varphi_{n,k\alpha} \psi_n - \psi_{n,k\alpha}^* \varphi_n^*) = 0; \\ \{\hat{b}, \hat{b}_{k\alpha}^\dagger\} = 0, &\Rightarrow \sum_n \psi_{n,k\alpha} \psi_n = 0.\end{aligned}\tag{31}$$

To compute the steady-state current (19) we need to express the current superoperator  $\hat{J}_\alpha$  in terms of quasiparticle superoperators. By doing so and taking into account property (29), we obtain the following expression for the steady-state current

$$\begin{aligned}J_\alpha &= \langle I| \hat{J}_\alpha | \rho \rangle \\ &= -2\text{Im} \sum_{m,k} t_{k\alpha} \psi_m \left( \varphi_{m,k\alpha} + \sum_n \psi_{n,k\alpha}^* F_{mn} \right),\end{aligned}\tag{32}$$

where  $F_{mn} = -i\langle I| \tilde{c}_n \hat{c}_m | \rho \rangle$  is the two-quasiparticle amplitude. Note, that this is an *exact* expression for the steady-state current. The only problem is to find the unknown  $F_{mn}$ .

In the absence of electron-phonon interaction, i.e., when the nonequilibrium density matrix does not contain quasiparticle excitations, the current through the central region



is given by the first terms in (32). In what follows we will refer to this current as a free-field current  $J_\alpha^{(0)}$ . The correction to the free-field current,  $\Delta J_\alpha$ , is given by the second term involving  $F_{mn}$ . In [13], it was shown how to compute  $F_{mn}$  making use of the perturbation theory. In the following subsections we demonstrate how to find  $F_{mn}$  within the NECI approach using different reference states for the density matrix expansion, namely, free-field and coherent reference states.

### 3.2. NECI expansion on free-field vacuum as a reference state

We define the free-field reference state as the steady-state density matrix in the absence of electron-phonon interaction, i.e.,  $|\rho^{(0)}\rangle = |\rho^{(0)}\rangle_{\text{el}} |\rho^{(0)}\rangle_{\text{ph}}$  and

$$L^{(0)} |\rho^{(0)}\rangle = 0. \quad (33)$$

The density matrix  $|\rho^{(0)}\rangle_{\text{el}}$  is a vacuum state for nonequilibrium quasiparticles, i.e.,

$$\hat{c}_n |\rho^{(0)}\rangle_{\text{el}} = \tilde{c}_n |\rho^{(0)}\rangle_{\text{el}} = 0. \quad (34)$$

To determine  $|\rho^{(0)}\rangle_{\text{ph}}$  we take into account the possibility that the phonon subsystem can contain a certain number of thermally excited vibrational quanta. Let  $N_\omega$  be the number of thermally excited phonons, i.e.,

$$N_\omega = \langle I | \hat{d}^\dagger \hat{d} | \rho^{(0)} \rangle_{\text{ph}}. \quad (35)$$

It is convenient to perform a non-unitary canonical transformation and introduce new phonon operators

$$\begin{aligned} \hat{\gamma} &= (1 + N_\omega) \hat{d} - N_\omega \tilde{d}^\dagger, \\ \hat{\gamma}^\dagger &= \hat{d}^\dagger - \tilde{d} \end{aligned} \quad (36)$$

and their tilde conjugated partners  $\tilde{\gamma}$ ,  $\tilde{\gamma}^\dagger$  such that  $|\rho^{(0)}\rangle_{\text{ph}}$  is the vacuum state for  $\hat{\gamma}$ ,  $\tilde{\gamma}$  superoperators,

$$\hat{\gamma} |\rho^{(0)}\rangle_{\text{ph}} = \tilde{\gamma} |\rho^{(0)}\rangle_{\text{ph}} = 0, \quad (37)$$

while  $\langle I |$  is the vacuum for  $\hat{\gamma}^\dagger, \tilde{\gamma}^\dagger$  superoperators (see property (A.7) in Appendix). Now, the free-field Liouvillian takes the form

$$L^{(0)} = \sum_n (\Omega_n \hat{c}_n^\dagger \hat{c}_n - \Omega_n^* \tilde{c}_n^\dagger \tilde{c}_n) + \omega_0 (\hat{\gamma}^\dagger \hat{\gamma} - \tilde{\gamma}^\dagger \tilde{\gamma}), \quad (38)$$

and the vacuum state  $|\rho^{(0)}\rangle$  for annihilation superoperators  $\hat{c}_n$ ,  $\tilde{c}_n$ ,  $\hat{\gamma}$ ,  $\tilde{\gamma}$  is the free-field reference state obeying condition (33). The free-field density matrix  $|\rho^{(0)}\rangle$  is normalized according to  $\langle I | \rho^{(0)} \rangle = 1$ .

Due to the electron-phonon interaction the exact steady-state density matrix contains multi-quasiparticle and multi-phonon excitations above the free-field density matrix. We are looking for the exact steady-state density matrix in the form

$$|\rho\rangle = (1 + S) |\rho^{(0)}\rangle, \quad (39)$$

where the operator  $S$  is given by the infinite power series of creation superoperators

$$S = \sum_{i=1}^{\infty} \alpha_i Q_i^\dagger, \quad Q_i^\dagger = \hat{c}_{m_1}^\dagger \dots \hat{c}_{m_k}^\dagger \tilde{c}_{n_1}^\dagger \dots \tilde{c}_{n_l}^\dagger (\gamma^\dagger)^p (\tilde{\gamma}^\dagger)^q. \quad (40)$$

Defined this way, the density matrix is normalized according to  $\langle I | \rho \rangle = 1$ , as  $\langle I |$  is a vacuum for quasiparticle and phonon creation superoperators. Since the density matrix is tilde-invariant (see the definition in Appendix A), the superoperator  $S$  obeys the property  $S = (S)^\sim$ , i.e. it remains the same if we complex conjugate all  $\alpha_i$  and replace the non-tilde superoperators by the tilde ones and vice versa.

We demand that the steady-state density matrix obeys the equation  $L | \rho \rangle = 0$ , therefore

$$\left\{ SL' + [L^{(0)} + L', S] \right\} | \rho^{(0)} \rangle = -L' | \rho^{(0)} \rangle. \quad (41)$$

This superoperator equation is equivalent to the infinite inhomogeneous system of linear equations for  $\alpha_i$  amplitudes in the expansion for nonequilibrium density matrix (40). Any truncation in the expansion (40) leads to approximate solution of equation (41).

Here, we consider the simplest form of  $S$  which allows us to calculate the correction for the free-field current. Namely,

$$S = i \sum_{mn} (F_{mn} + Z_{mn} \hat{\gamma}^\dagger + Z_{nm}^* \tilde{\gamma}^\dagger) \hat{c}_m^\dagger \tilde{c}_n^\dagger + W (\hat{\gamma}^\dagger + \tilde{\gamma}^\dagger), \quad (42)$$

where  $F_{nm} = F_{mn}^*$  and  $W$  is a real number. Inclusion of  $W$  and  $Z$  terms into the density matrix expansion is necessary, since  $\hat{c}_m^\dagger \tilde{c}_n^\dagger \hat{\gamma}^\dagger$ ,  $\tilde{\gamma}^\dagger$  configurations and their tilde-conjugate contribute to the right-hand side of (41) (see the expression for  $L'$  below). If we neglect them, we observe a homogeneous linear system having a trivial solution. In this sense, these terms are correlations inducing terms.

In order to find equations for the amplitudes  $F$ ,  $Z$ ,  $W$  we first express  $L'$  in terms of nonequilibrium quasiparticle superoperators:

$$\begin{aligned} L' = & \kappa \sum_{mn} \left\{ [L_{mn}^{(1)} \hat{\gamma}^\dagger + L_{mn}^{(2)} \tilde{\gamma}^\dagger + L_{mn}^{(3)} (\gamma + \tilde{\gamma})] \hat{c}_m^\dagger \tilde{c}_n - \text{t.c.} \right\} \\ & - i \kappa \sum_{mn} [L_{mn}^{(4)} \hat{\gamma}^\dagger - (L_{nm}^{(4)})^* \tilde{\gamma}^\dagger + L_{mn}^{(5)} (\gamma + \tilde{\gamma})] \hat{c}_m^\dagger \tilde{c}_n^\dagger \\ & - i \kappa \sum_{mn} L_{mn}^{(6)} (\hat{\gamma}^\dagger - \tilde{\gamma}^\dagger) \hat{c}_m \tilde{c}_n + \kappa n^{(0)} (\hat{\gamma}^\dagger - \tilde{\gamma}^\dagger). \end{aligned} \quad (43)$$

Here the coefficients  $L_{mn}^{(p)}$  are

$$\begin{aligned} L_{mn}^{(1)} &= [(\psi_m - \varphi_m) + N_\omega \psi_m] \psi_n, \\ L_{mn}^{(2)} &= [\varphi_m + N_\omega \psi_m] \psi_n, \quad L_{mn}^{(3)} = \psi_m \psi_n, \\ L_{mn}^{(4)} &= [(\psi_m - \varphi_m) \varphi_n^* + N_\omega (\psi_m \varphi_n^* - \varphi_m \psi_n^*)] \\ L_{mn}^{(5)} &= \psi_m \varphi_n^* - \varphi_m \psi_n^*, \quad L_{mn}^{(6)} = \psi_m \psi_n^*, \end{aligned} \quad (44)$$

and  $n^{(0)} = \langle I | \hat{a}^\dagger \hat{a} | \rho^{(0)} \rangle = \sum_n \psi_n \varphi_n$  is the free-field population of the electron level. Note, that after normal ordering  $L'$  does not involve terms containing annihilation superoperators only. Therefore, the condition  $\langle I | L = 0$  is fulfilled.

Substituting  $S$  given by (42) into (41) and demanding equation (41) to be fulfilled up to terms included into  $S$  we derive the system of linear equations for unknown amplitudes:

$$\begin{aligned}
 & F_{mn}(\Omega_m - \Omega_n^*) + \kappa \sum_i L_{mi}^{(3)} [Z_{in} + Z_{ni}^*] \\
 & - \kappa \sum_i (L_{ni}^{(3)})^* [Z_{mi} + Z_{im}^*] - 2\kappa L_{mn}^{(5)} W = 0 \\
 & Z_{mn}(\Omega_m - \Omega_n^* + \omega_0) + \kappa n^{(0)} F_{mn} \\
 & + \kappa \sum_i [L_{mi}^{(1)} F_{in} - (L_{ni}^{(2)})^* F_{mi}] = \kappa L_{mn}^{(4)} \\
 & W\omega_0 - \kappa \sum_{mn} L_{mn}^{(6)} F_{mn} = -\kappa n^{(0)}.
 \end{aligned} \tag{45}$$

It should be pointed out that the first and the last equations above are exact in the sense that the inclusion of other terms into  $S$  does not modify these equations.

### 3.3. Coherent reference state for NECI expansion

Within NECI method we have some flexibility in the choice of the reference state. Ideally, we would like to put as much correlations as possible into the reference state while maintaining the possibility to define it as a vacuum for some quasiparticle annihilation operators. We note that  $L'$  given by (43) contains the term  $\kappa n^{(0)}(\hat{\gamma}^\dagger - \tilde{\gamma}^\dagger)$  which is linear in phonon superoperators. It gives us an idea to eliminate this term by the non-unitary canonical transformation

$$\hat{\xi}^\dagger = \hat{\gamma}^\dagger, \quad \hat{\xi} = \hat{\gamma} + \kappa \frac{n^{(0)}}{\omega_0} \tag{46}$$

and  $\tilde{\xi}^\dagger = (\hat{\xi}^\dagger)^\sim$ ,  $\tilde{\xi} = (\hat{\xi})^\sim$ . The vacuum of the "shifted" phonon operators is the coherent state

$$|\rho^{(*)}\rangle = \exp \left\{ -\kappa \frac{n^{(0)}}{\omega_0} (\gamma^\dagger + \tilde{\gamma}^\dagger) \right\} |\rho^{(0)}\rangle, \tag{47}$$

normalized according to  $\langle I | \rho^{(*)} \rangle = 1$ .

Let us consider this coherent state as a reference state for the configuration interaction expansion, i.e.

$$|\rho\rangle = (1 + S) |\rho^{(*)}\rangle, \tag{48}$$

where  $S$  is given by

$$S = \sum_{i=1}^{\infty} \beta_i Q_i, \quad Q_i = c_{m_1}^\dagger \dots c_{m_k}^\dagger \tilde{c}_{n_1}^\dagger \dots \tilde{c}_{n_l}^\dagger (\xi^\dagger)^p (\tilde{\xi}^\dagger)^q. \tag{49}$$

To distinguish from the NECI expansion on the free-field reference state, we denote the present method as NECI\*. The advantage of NECI\* approach is that it effectively includes multi-phonon excitation and de-excitation processes via the exponent (47) in the coherent reference state.

To fulfill the condition  $L(1 + S)|\rho^{(*)}\rangle = 0$ , the superoperator  $S$  must obey the equation

$$\{SL' + [L^{(0)} + L', S]\}|\rho^{(*)}\rangle = -L'|\rho^{(*)}\rangle, \quad (50)$$

where

$$L^{(0)} = \sum_n (\Omega_n c_n^\dagger c_n - \Omega_n^* \tilde{c}_n^\dagger \tilde{c}_n) + \omega_0 (\xi^\dagger \xi - \tilde{\xi}^\dagger \tilde{\xi}), \quad (51)$$

and

$$\begin{aligned} L' = & -2\kappa^2 \frac{n^{(0)}}{\omega_0} \sum_{mn} \left\{ [L_{mn}^{(3)} c_m^\dagger c_n - \text{t.c.}] - iL_{mn}^{(5)} c_m^\dagger \tilde{c}_n^\dagger \right\} \\ & + \kappa \sum_{mn} \left\{ [L_{mn}^{(1)} \xi^\dagger + L_{mn}^{(2)} \tilde{\xi}^\dagger + L_{mn}^{(3)} (\xi + \tilde{\xi})] c_m^\dagger c_n - \text{t.c.} \right\} \\ & - i\kappa \sum_{mn} [L_{mn}^{(4)} \xi^\dagger - (L_{nm}^{(4)})^* \tilde{\xi}^\dagger + L_{mn}^{(5)} (\xi + \tilde{\xi})] c_m^\dagger \tilde{c}_n^\dagger \\ & - i\kappa \sum_{mn} L_{mn}^{(6)} (\xi^\dagger - \tilde{\xi}^\dagger) c_m \tilde{c}_n. \end{aligned} \quad (52)$$

If all terms are included into the density matrix expansion, the two methods, NECI and NECI\*, coincide because both are formally exact. However, when we truncate the expansion for the density matrixes, NECI and NECI\* give different results.

Likewise to NECI, we take the operator  $S$  in the form

$$S = i \sum_{mn} (F_{mn} + Z_{mn} \xi^\dagger + Z_{nm}^* \tilde{\xi}^\dagger) c_m^\dagger \tilde{c}_n^\dagger + W(\xi^\dagger + \tilde{\xi}^\dagger). \quad (53)$$

Demanding that equation (50) is fulfilled up to terms included into  $S$  we obtain the following system of equations

$$\begin{aligned} & F_{mn}(\Omega_m - \Omega_n^*) - \frac{2\kappa^2 n^{(0)}}{\omega_0} \sum_i [L_{mi}^{(3)} F_{in} - (L_{ni}^{(3)})^* F_{mi}] \\ & + \kappa \sum_i L_{mi}^{(3)} [Z_{in} + Z_{ni}^*] - \kappa \sum_i (L_{ni}^{(3)})^* [Z_{mi} + Z_{im}^*] \\ & - 2\kappa L_{mn}^{(5)} W = -\frac{2\kappa^2 n^{(0)}}{\omega_0} L_{mn}^{(5)} \\ & Z_{mn}(\Omega_m - \Omega_n^* + \omega_0) - \frac{2\kappa^2 n^{(0)}}{\omega_0} \sum_i [L_{mi}^{(3)} Z_{in} - (L_{ni}^{(3)})^* Z_{mi}] \\ & + \kappa \sum_i [L_{mi}^{(1)} F_{in} - (L_{ni}^{(2)})^* F_{mi}] + \frac{2\kappa^2 n^{(0)}}{\omega_0} L_{mn}^{(5)} W = \kappa L_{mn}^{(4)} \\ & W\omega_0 - \kappa \sum_{mn} L_{mn}^{(6)} F_{mn} = 0. \end{aligned} \quad (54)$$

Solving this system we compute the two-quasiparticle amplitudes  $F_{mn}$  and, hence, obtain the NECI\* correction to the free-field current. Here again, the first and the last equations are exact, i.e., additions of new terms to  $S$  does not modify these equations.

### 3.4. NECI is a current-conserving theory

The truncation of the density matrix expansion introduces approximations into the theory. It is important to ensure that observables computed with the truncated density matrix still satisfy conservation laws dictated by the symmetries of the underlying Hamiltonian.[19, 20, 21] In the nonequilibrium case it is particularly important to demonstrate that the proposed configuration interaction theory preserves the particle number continuity equation in all orders of configurations included into the density matrix. To this end, we are going to prove that there is no artificial current leakage from the system introduced by the approximations and the current which enters the system from the left reservoir is exactly the same as the current which leaves the system to the right reservoir:

$$J_L + J_R = 0. \quad (55)$$

Let us first prove the current conservation in the free-field approximation. Indeed, the following equality is true

$$\begin{aligned} J_L^{(0)} + J_R^{(0)} &= -2\text{Im} \sum_{n,k\alpha} t_{k\alpha} \varphi_{n,k\alpha} \psi_n \\ &= -\text{Im} \sum_{n,k\alpha} t_{k\alpha} (\varphi_{n,k\alpha} \psi_n - \psi_{n,k\alpha} \varphi_n) \\ &= \text{Im} \sum_{n,k\alpha} t_{k\alpha} f_{k\alpha} \psi_{n,k\alpha} \psi_n = 0. \end{aligned} \quad (56)$$

Here, we have used relations (31) as well as the first equations in (27) and (28).

The solution of system (45) (or (54)) provides us the two-quasiparticle amplitudes  $F_{mn}$  and the NECI (NECI\*) correction  $\Delta J_\alpha$  to the free-field current (see equation (32)). With the help of the first equation in (27) and using the property  $F_{nm} = F_{mn}^*$  we find

$$\begin{aligned} \Delta J_L + \Delta J_R &= -2\text{Im} \sum_{mn,k\alpha} t_{k\alpha} \psi_{n,k\alpha}^* \psi_m F_{mn} \\ &= -2\text{Im} \sum_{mn} \psi_m \psi_n^* (\varepsilon_0 - \Omega_n^*) F_{mn} \\ &= -\text{Im} \sum_{mn} \psi_m \psi_n^* (\Omega_m - \Omega_n^*) F_{mn}. \end{aligned} \quad (57)$$

Expressing  $(\Omega_m - \Omega_n^*) F_{mn}$  from the first *exact* equation in (45) (or (54)) and taking into account the explicit expressions for  $L_{mn}^{(3)}$ ,  $L_{mn}^{(5)}$  we find that  $\Delta J_L + \Delta J_R = 0$ . Note that this result does not depend on the particular choice of  $S$ , i.e. it is valid for any truncated NECI (or NECI\*) expansion.

Thus, the presented configuration interaction theory is current-conserving in all orders of nonequilibrium quasiparticle configurations included into the density matrix expansion.

## 4. Numerical Results

In our numerical calculations we assume that the applied voltage  $V$  symmetrically shifts the on-site energies, i.e.,  $\epsilon_{L,R} = \pm 0.5V$ . Additionally, we assume that the electrodes are half filled, i.e., the corresponding left and right chemical potentials are positioned at  $\epsilon_{L,R}$ . We choose the following parameters of the electrodes:  $V_L = V_R = 2.5$ ,  $t_L = t_R = 1.0$  and the temperature is  $T = 0.1$ . Since the on-site energies  $\epsilon_{L,R}$  are affected by the applied voltage the left and right electrodes are nonidentical. In the subsequent discussion we set the applied voltage  $V = 1.0$ .

For the buffer zones we take a finite chain of  $N$  atoms from each electrode. Therefore the energy spectrum of each buffer zone is given by

$$\varepsilon_{k\alpha} = \epsilon_\alpha + 2V_\alpha \cos\left(\frac{\pi k}{N+1}\right), \quad k = 1, \dots, N. \quad (58)$$

and the tunneling matrix elements in (11) are

$$t_{k\alpha} = t_\alpha \sqrt{\frac{2}{N+1}} \sin\left(\frac{\pi k}{N+1}\right). \quad (59)$$

The parameter  $\gamma_{k\alpha}$  in Lindblad operators (15) is taken to be equal the distance between neighbor energy levels in the buffer zones, i.e.,  $\gamma_{k\alpha} = \varepsilon_{k\alpha} - \varepsilon_{k+1\alpha}$ . In our calculations we include  $N = 800$  sites into each buffer zone. This size of the buffer zone has been proven to give the exact results for the steady state density matrix of the system. [14, 15]

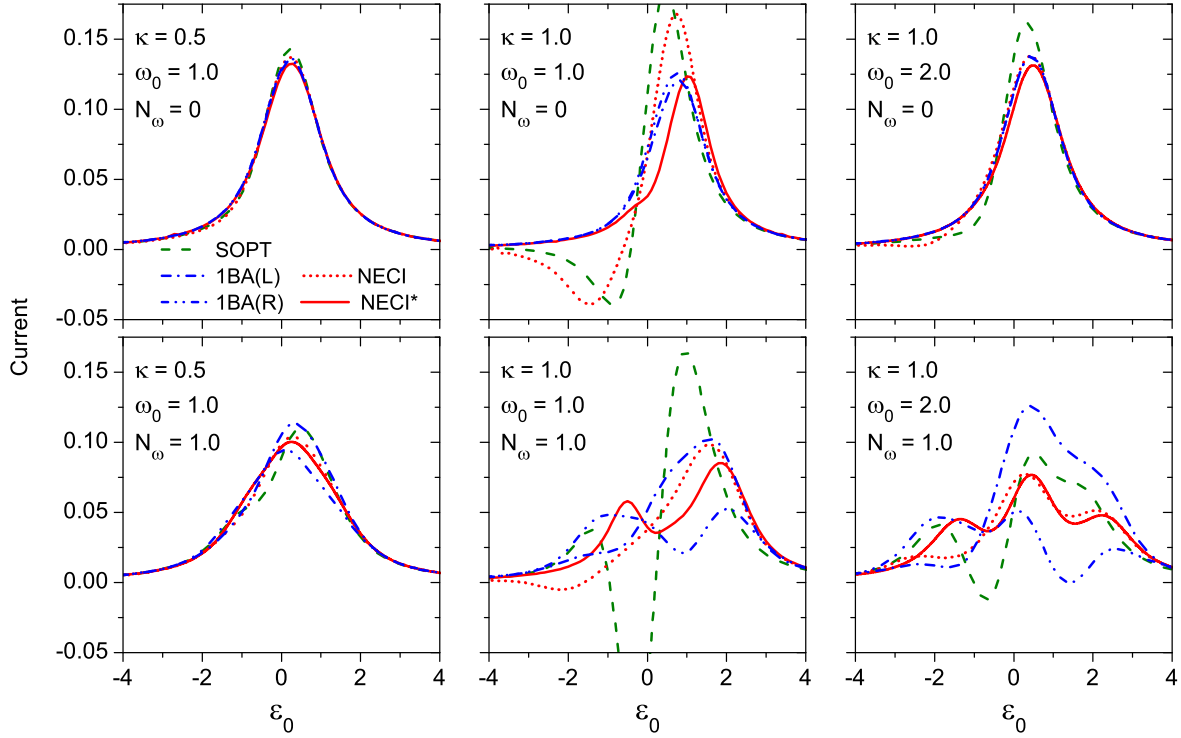
Let us say some words concerning numerical solution of the systems of equations (45) and (54). To be specific we consider the system obtained within NECI theory (for the NECI\* system we have used the same solution method). The dimension of the system is of the order of  $4M^2$ , where  $M = 2N+1$ . Although the systems is sparse, it contains about  $12M^3$  nonzero matrix elements. Assuming that each complex number requires 16 bytes of computer memory, we find that for  $N = 800$  the total system matrix needs about 700 gigabyte for storage. A required memory size can be reduced drastically if we contract the indices by introducing the following linear combinations

$$f_n = \sum_m F_{mn}, \quad z_n = \sum_m (Z_{mn} + Z_{nm}^*) \quad (60)$$

and their complex conjugate. Note, that the correction to the free-field current can be written as

$$\Delta J_\alpha = - \sum_{n,k} t_{k\alpha} \psi_{n,k\alpha}^* f_n. \quad (61)$$

Then, it is possible to rewrite the system (45) as a system for  $f_n$ ,  $z_n$  and their complex conjugate. The obtained system contains  $4M + 1$  equations and the total number of



**Figure 2.** Current through the central region calculated within different approaches as a function of the level energy,  $\varepsilon_0$ .

nonzero elements is about  $6M^2$ . To solve the system we have used standard routines from Intel MKL Fortran library.

In figure 2 we compare the second-order perturbation theory (SOPT) and first Born approximation (1BA) results for the electron current with the results obtained within NECI and NECI\* method for different values of the central region parameters. The currents are shown as functions of the level energy,  $\varepsilon_0$ , i.e., for different transport regimes (resonant/off-resonant). Since the first Born approximation does not guarantee the current conservation, we plot the 1BA current from the left electrode to the central region as well as the 1BA current from the central region to the right electrode. The details of SOPT and 1BA calculations using nonequilibrium Green's functions are presented in Appendix B.

We first consider the case when  $N_\omega = 0$ , i.e., there is no equilibrium thermally excited vibrational quanta. As evident from the figure in this case the first Born approximation does not violate the current conservation significantly since the left and right currents are close to each other. We see that for a weak coupling ( $\kappa = 0.5$ ) all approaches predict a similar  $\varepsilon_0$  dependency of the current, which reaches a maximum value at  $\varepsilon_0 \approx \kappa^2/\omega_0$ . The peak value of the SOPT and 1BA currents slightly exceeds the NECI and NECI\* ones. When we increase the electron-phonon coupling ( $\kappa = 1.0$ ) both SOPT and NECI currents become unphysical negative in the off-resonant regime when the electronic level  $\varepsilon_0$  is below the electrode chemical potentials. In the resonant regime

SOPT and NECI currents significantly overestimate the results of other approaches. In addition, comparing NECI\*-based calculations with other calculations, we can see that NECI\* approach gives the current peak position at  $\varepsilon_0 \approx \kappa^2/\omega_0$ . If we now consider the case of larger phonon energy ( $\omega_0 = 2.0$ ) we notice that negative SOPT and NECI currents in the off-resonant regime disappear and all approaches again demonstrate a similar  $\varepsilon_0$  dependency of the current.

Now we consider results obtained for nonzero number of thermally excited equilibrium vibrational quanta. Namely, we assume that  $N_\omega = 1.0$ . In this case the first Born approximation clearly reveals its current nonconserving nature. This is illustrated in the lower panels of figure 2 where we can see that the left and right 1BA currents can be very different from each other. Nevertheless, in the weak electron-phonon coupling regime ( $\kappa = 0.5$ ), all approaches predict qualitatively similar dependence of the current upon the electronic level  $\varepsilon_0$ . Comparing the result for  $\kappa = 0.5$ ,  $N_\omega = 1.0$  with those for  $\lambda = 0.5$ ,  $N_\omega = 0$  we notice that inclusion of thermally excited equilibrium vibrational quanta into consideration produces slightly broadened current peak with a reduced amplitude. When the coupling constant increases ( $\kappa = 1.0$ ) SOPT and NECI approaches again show unphysical negative current in the off-resonant regime when  $\varepsilon_0$  is below the electrode chemical potentials. Moreover, for  $\kappa = 1.0$  the electron-phonon coupling splits the NECI\* current peak, while NECI approach gives only one peak. For a larger phonon energy,  $\omega_0 = 2.0$ , NECI\* predicts three pronounced peaks, while NECI gives only two peaks. In addition, SOPT and 1BA show unphysically negative current for certain values of  $\varepsilon_0$ .

From the above consideration it is evident that the configuration interaction method built on the coherent reference state is preferable when the electron-phonon coupling is large or when there are thermally excited vibrational quanta in the system. This result is not surprising since, contrary to NECI, NECI\* method accounts for multi-photon excitations and de-excitation processes which are important in these regimes.

## 5. Conclusions

We developed nonequilibrium configuration interaction method, which formally gives the exact solution of out-of-equilibrium correlated many-body problem. Our approach is based on a superoperator representation of the Lindblad master equation for the reduced density matrix of the embedded quantum system. It was shown that the steady-state density matrix can be decomposed in Liouville-Fock space in terms of nonequilibrium multi-quasiparticle excitations above the reference vacuum state. The amplitudes of these excitations provide a measure of the many-body nonequilibrium correlations.

The theory was applied to study the inelastic electron transport through the system with electron-phonon interaction. To compute the current we used truncated expansion of the steady-state density matrix. Two different reference states were considered: free-field vacuum and coherent state. It was proved that both approximations are current conserving in all orders of the density matrix expansion. The current through the



system was computed for different model parameters and compared with the second-order perturbation theory and the first Born approximation results. It was shown that the configuration interaction method based on the coherent reference state is superior to the other approaches when the electron-phonon coupling is large or when there are thermally excited vibrational quanta in the system.

The method can be readily extended to dynamical nonequilibrium case by making the amplitudes in the density matrix expansion time-dependent functions.

## Appendix A. Brief overview of the superoperator formalism

Here we briefly review the formalism of superoperators acting in the Liouville-Fock space, while the detailed discussion is given in [9, 13]. We suppose that the Fock space of the system under consideration is spanned by the complete orthonormal set of basis vectors  $|m\rangle = |m_1, m_2, \dots\rangle$  which are eigenvectors of the particle number operator:

$$a_k^\dagger a_k |m\rangle = m_k |m\rangle, \quad \sum_m |m\rangle\langle m| = I, \quad \langle n|m\rangle = \delta_{nm}. \quad (\text{A.1})$$

The operators in the Fock space form themselves a linear vector space called the Liouville-Fock space. The set of vectors  $|mn\rangle \equiv |m\rangle\langle n|$  constitutes a orthonormal basis in the Liouville-Fock space. Thus, every Fock space operator  $A = \sum_{mn} A_{mn} |m\rangle\langle n|$  can be considered as a Liouville-Fock space ket-vector  $|A\rangle = \sum_{mn} A_{mn} |mn\rangle$ . The adjoint operator  $A^\dagger$  is represented by the bra-vector  $\langle A|$ .

The scalar product in the Liouville-Fock space is defined as  $\langle A_1|A_2\rangle = \text{Tr}(A_1^\dagger A_2)$ . In particular, the scalar product of a vector  $|A\rangle$  with  $\langle I|$  is equivalent to the trace operation in the Fock space,  $\langle I|A\rangle = \text{Tr}(A)$ .

Now we introduce creation and annihilation superoperators. Fermion creation and annihilation superoperators are defined as

$$\begin{aligned} \hat{a}_k |mn\rangle &\equiv a_k |m\rangle\langle n|, \quad \tilde{a}_k |mn\rangle \equiv i(-1)^\mu |m\rangle\langle n| a_k^\dagger \\ \hat{a}_k^\dagger |mn\rangle &\equiv a_k^\dagger |m\rangle\langle n|, \quad \tilde{a}_k^\dagger |mn\rangle \equiv i(-1)^\mu |m\rangle\langle n| a_k, \end{aligned} \quad (\text{A.2})$$

where  $\mu = \sum_k (m_k + n_k)$ . For bosonic creation and annihilation superoperators we drop the phase factor  $i(-1)^\mu$ . So defined superoperators satisfy the same (anti)commutation relations as their Fock space counterparts. Additionally, the basis vectors of the Liouville-Fock space are super-particle number eigenvectors, i.e.,

$$\hat{a}_k^\dagger \hat{a}_k |mn\rangle = m_k |mn\rangle, \quad \tilde{a}_k^\dagger \tilde{a}_k |mn\rangle = n_k |mn\rangle. \quad (\text{A.3})$$

For an operator  $A = A(a^\dagger, a)$  we formally define two superoperators

$$\hat{A} = A(\hat{a}^\dagger, \hat{a}), \quad \tilde{A} = A^*(\tilde{a}^\dagger, \tilde{a}) \quad (\text{A.4})$$

and refer to them as non-tilde and tilde superoperators, respectively. The connection between non-tilde and tilde superoperators is given by the "tilde conjugation rules"

$$\begin{aligned} (c_1 \hat{A}_1 + c_2 \hat{A}_2) \tilde{\gamma} &= c_1^* \tilde{A}_1 + c_2^* \tilde{A}_2, \\ (\hat{A}_1 \hat{A}_2) \tilde{\gamma} &= \tilde{A}_1 \tilde{A}_2, \quad (\tilde{A}) \tilde{\gamma} = \hat{A}, \quad (\hat{A}^\dagger) \tilde{\gamma} = (\tilde{A})^\dagger. \end{aligned} \quad (\text{A.5})$$

With the help of superoperators an arbitrary Liouville-Fock space vector can be represented as

$$|A\rangle = \hat{A} |I\rangle = \sigma_A \tilde{A}^\dagger |I\rangle. \quad (\text{A.6})$$

Hereinafter, the phase  $\sigma_A = -i$  if  $A$  is a fermionic operator and  $\sigma_A = +1$  if  $A$  is a bosonic operator. We also define a Liouville-Fock space vector tilde conjugated to a given one,  $|A\tilde{\rangle} \equiv \tilde{A} |I\rangle = \sigma_A^* \hat{A}^\dagger |I\rangle$ . Therefore, if  $A$  is a Hermitian bosonic operator then  $|A\rangle$  is tilde-invariant, i.e.,  $|A\rangle = |A\tilde{\rangle}$ . The examples of tilde-invariant vectors are  $|I\rangle$  and the density matrix  $|\rho\rangle = \hat{\rho} |I\rangle$ .

Considering the adjoint of (A.6) and assuming that  $\hat{A} = \hat{a}$  or  $\hat{a}^\dagger$  we find

$$\langle I | (\hat{a}^\dagger - \sigma_a^* \tilde{a}) = \langle I | (\tilde{a}^\dagger - \sigma_a \hat{a}) = 0. \quad (\text{A.7})$$

This gives us the idea introduce superoperator  $\hat{b}^\dagger = \hat{a}^\dagger - \sigma_a^* \tilde{a}$  and its tilde conjugate,  $\tilde{b}^\dagger = \tilde{a}^\dagger - \sigma_a \hat{a}$ , which annihilate the bra-vector  $\langle I |$ .

For the product of operators the following relation is valid

$$|A_1 A_2\rangle = \hat{A}_1 |A_2\rangle = \tau \tilde{A}_2^\dagger |A_1\rangle, \quad (\text{A.8})$$

where  $\tau = i$  if both  $A_1$  and  $A_2$  are fermionic and  $\tau = \sigma_{A_2}$  otherwise. With the help of relations (A.8) the average of an operator  $A$  in the state with the density matrix  $\rho$  can be calculated as the matrix element of the corresponding superoperator  $\hat{A}$  sandwiched between  $\langle I |$  and  $|\rho\rangle$ :

$$\langle A \rangle = \text{Tr}(A\rho) = \langle I | A\rho \rangle = \langle I | \hat{A} |\rho \rangle. \quad (\text{A.9})$$

Let us now consider the Lindblad master equation (12). In the Liouville-Fock space this equation takes the form

$$i \frac{d}{dt} |\rho(t)\rangle = |H\rho(t)\rangle - |\rho(t)H\rangle + i |\Pi\rho(t)\rangle. \quad (\text{A.10})$$

Applying relations (A.8), i.e., taking into account that  $|H\rho(t)\rangle = \hat{H} |\rho(t)\rangle$ ,  $|\rho(t)H\rangle = \tilde{H} |\rho(t)\rangle$ ,  $|a_{k\alpha}\rho(t)a_{k\alpha}^\dagger\rangle = \hat{a}_{k\alpha} |\rho(t)a_{k\alpha}^\dagger\rangle = i\hat{a}_{k\alpha}\tilde{a}_{k\alpha} |\rho(t)\rangle$ , etc., we can rewrite this equation in the Schrödinger-like form (16). The time evolution of the density matrix is governed by the non-Hermitian Liouville superoperator  $L$ . Two important properties of  $L$  should be noted: 1. Since the density matrix is tilde invariant, then  $(L\tilde{\phantom{x}}) = -L$ ; 2. Taking the time derivative of  $\langle I | \rho(t) \rangle = 1$  we find  $\langle I | L = 0$ , i.e.,  $\langle I |$  is the left zero-eigenvalue eigenvector of  $L$ .

## Appendix B. Details of NEGF calculations

In this Appendix we briefly describe the details of transport calculations based on nonequilibrium Green's functions relevant to our discussion. We first define four kind of interacting Green's functions:  $G^{r,a}(\omega)$  and  $G^{\lessgtr}(\omega)$ . The retarded and advanced Green's functions are given as the solutions of Dyson's equation

$$G^r = G_0^r + G_0^r \Sigma_{\text{int}}^r G^r, \quad G^a = (G^r)^*, \quad (\text{B.1})$$

where  $G_0^r = [\omega - \varepsilon_0 - \Sigma_L^r - \Sigma_R^r]^{-1}$  is the noninteracting retarded Green's function. The lesser and greater Green's functions satisfy the Keldysh equation

$$G^{\lessgtr} = G^r(\Sigma_L^{\lessgtr} + \Sigma_R^{\lessgtr} + \Sigma_{\text{int}}^{\lessgtr})G^a. \quad (\text{B.2})$$

The interaction self-energies  $\Sigma_{\text{int}}^{\lessgtr,r}$  account for the electron-phonon coupling in the central region, while the electrode self-energies,  $\Sigma_{\alpha}^{\lessgtr,r}$ , describe the effects of the coupling between the central region and the electrodes

$$\begin{aligned} \Sigma_{\alpha}^{<}(\omega) &= +2i\text{Im}[\Sigma_{\alpha}^r(\omega)](f_{\alpha}(\omega) - 1), \\ \Sigma_{\alpha}^{>}(\omega) &= +2i\text{Im}[\Sigma_{\alpha}^r(\omega)]f_{\alpha}(\omega). \end{aligned} \quad (\text{B.3})$$

Here,  $f_{\alpha}$  is the Fermi-Dirac distribution function with chemical potential  $\mu_{\alpha}$  and temperature  $T$ .

Once the full interacting Green's functions are defined, we can write explicit expressions for physical quantities of interest. For the net electric current passing through the  $\alpha$  electrode to the central region we adopt the Meir-Wingreen transport formula [22]

$$J_{\alpha} = \int \frac{d\omega}{2\pi} \{ \Sigma_{\alpha}^{<}(\omega) G^{>}(\omega) - \Sigma^{>}(\omega) G^{<}(\omega) \}, \quad (\text{B.4})$$

and for the electron level nonequilibrium population we use

$$n_{\text{el}} = \int \frac{d\omega}{2\pi i} G^{<}(\omega). \quad (\text{B.5})$$

Neglecting the electron-phonon interaction, i.e., assuming  $\Sigma_{\text{int}}^{\lessgtr,r} = 0$ , we obtain the free-field current,  $J_{\alpha}^{(0)}$ , and electron populations,  $n_{\text{el}}^{(0)}$ .

The exact electron-phonon interaction self-energies  $\Sigma_{\text{int}}^{\lessgtr,r}$  in Green's functions (B.2) and (B.1) contain all possible diagrams that satisfy Feynman rules. In this work, we calculate  $\Sigma_{\text{int}}^{\lessgtr,r}$  from the lowest order self-energy diagrams referred to as the Hartree and Fock diagrams,  $\Sigma_{\text{int}}^{\lessgtr,r} = \Sigma_H^{\lessgtr,r} + \Sigma_F^{\lessgtr,r}$ . This approach is known as the first Born approximation (1BA). Using the standard rules of Feynman's diagrams and the Langreth theorem for analytical continuation we find the Hartree and Fock self-energies

$$\begin{aligned} \Sigma_H^{\lessgtr,r}(\omega) &= -i\kappa^2 D_0^{\lessgtr,r}(\omega = 0) \int \frac{d\omega'}{2\pi} G_0^{<}(\omega'), \\ \Sigma_F^{\lessgtr}(\omega) &= i\kappa^2 \int \frac{\omega'}{2\pi} D_0^{\lessgtr}(\omega') G_0^{\lessgtr}(\omega - \omega'), \\ \Sigma_F^r(\omega) &= i\kappa^2 \int \frac{\omega'}{2\pi} \left\{ D_0^{<}(\omega - \omega') G_0^r(\omega') \right. \\ &\quad \left. + D_0^r(\omega - \omega') [G_0^{<}(\omega') + G_0^r(\omega')] \right\}. \end{aligned} \quad (\text{B.6})$$

With the usual definition for the bare phonon Green's functions

$$\begin{aligned} D_0^{\lessgtr}(\omega) &= -2\pi i [N_{\omega} \delta(\omega \mp \omega_0) + (N_{\omega} + 1) \delta(\omega \pm \omega_0)] \\ D_0^r(\omega) &= \frac{1}{\omega - \omega_0 + i0^+} - \frac{1}{\omega + \omega_0 + i0^+}, \end{aligned} \quad (\text{B.7})$$

where  $N_\omega$  is the equilibrium occupation number of the phonon mode with frequency  $\omega_0$ . We see that  $\Sigma_H^\lessgtr = 0$  because  $D_0^\lessgtr(\omega = 0) = 0$  unless  $\omega_0 = 0$ . Since  $D_0^r(\omega = 0) = -2/\omega_0$ , we have  $\Sigma_H^r(\omega) = -2\kappa^2 n_{\text{el}}^{(0)}/\omega_0$ .

Substituting self-energies (B.6) into Dyson equation (B.1) and Keldysh equation (B.2) we find the lesser and greater Green's functions within the first Born approximation. Although the 1BA does not generally guarantee current conservation, computationally it is not as demanding as the self-consistent Born approximation when the full interacting Green's functions are used in the definition of the self-energies (B.6).

In the case of weak electron-phonon coupling the second-order perturbation theory (SOPT) may be a good approximation. In this approximation the lesser and greater Green's functions obtained within 1BA are expanded with respect to electron-phonon coupling up to  $\kappa^2$  terms

$$G^\lessgtr(\omega) \approx G_0^\lessgtr(\omega) + G_0^r(\omega)\Sigma_{\text{int}}^\lessgtr(\omega)G_0^a(\omega) + 2G_0^\lessgtr(\omega)\text{Re}[G_0^r(\omega)\Sigma_{\text{int}}^r(\omega)]. \quad (\text{B.8})$$

Substituting this expression into (B.4) we evaluate the current within the second-order perturbation theory. It should be noted that this approximation satisfies the current conservation condition.

In the end of this Appendix we present explicit expression for the electrode retarded self-energy. For the semi-infinite one-dimensional chain with on-site energy  $\epsilon_\alpha$  and hopping parameter  $V_\alpha$  coupled to the central region with hopping matrix element  $t_\alpha$ , the retarded self-energy  $\Sigma_\alpha^r(\omega) = \Lambda_\alpha(\omega) - 0.5i\Gamma_\alpha(\omega)$  can be written in an analytical form as

$$\Lambda_\alpha(\omega) = \frac{\Gamma_{0\alpha}}{2} \begin{cases} x, & |x| \leq 1 \\ x - \text{sgn}(x)\sqrt{x^2 - 1}, & |x| > 1, \end{cases} \\ \Gamma_\alpha(\omega) = \Gamma_{0\alpha}\Theta(1 - |x|)\sqrt{1 - x^2}, \quad (\text{B.9})$$

where  $x = (\omega - \epsilon_\alpha)/(2|V_\alpha|)$ ,  $\Gamma_{0\alpha} = 2t_\alpha^2/|V_\alpha|$ .

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